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Title: SYSTEMATIC STUDY OF ANISTROPIC PROPERTIES OF CERIUM NICKEL GERMANIUM (2)

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Systematic study of anisotropic properties of $CeNiGe_2$

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We have studied its anisotropic properties by the measurements of electrical resistivity, magnetic susceptibility and magnetization on a single crystal of $CeNiGe_2$ with a layered crystal structure. It is confirmed that $CeNiGe_2$ undergoes two-step antiferromagnetic transition at T_N^{I} = 4 K and T_N^{II} = 3 K as reported earlier on polycrystalline samples. $CeNiGe_2$ is found to exhibit highly anisotropic properties with an easy magnetization axis along the longest crystallographic b direction. The magnetization ratio $M(H//b)/M(H\perp b)$ is estimated to be about 15 at 5 T. The resistivity ratio $\rho_{//b}/\rho_{\perp b}$ increases from 5 to 65 on heating from 2 to 200 K. The in-plane resistivity $\rho_{//b}(T)$ shows double maxima typical of that expected when an interplay of crystal-field and Kondo effects plays a role.

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Ce-based intermetallics are of fundamental importance for the understanding of their various ground states. The type of magnetic ground state in heavy-fermion compounds is controlled by the competition between the indirect RKKY exchange (T_{RKKY}) and on-site Kondo (T_K) interactions. For the strong-hybridization limit ($T_K > T_{RKKY}$), the ground state is nonmagnetic as found in $CeCu_6$.[1] In the limit of medium to week hybridization ($T_K \approx T_{RKKY}$), different ground states can be found. CePdSn is a typical Kondo antiferromagnet with $T_N = 7$ K,[2] while $Ce_2Ni_3Ge_5$ exhibits two magnetic phase transitions at $T_{N1} = 5.1$ K and $T_{N2} = 4.5$ K.[3] For the weak-hybridization limit ($T_K < T_{RKKY}$), the system orders magnetically.

In the present paper, we report magnetization, magnetic susceptibility, specific heat, and electrical resistivity measurements on a single crystal of $CeNiGe_2$. This material crystallizes in the orthorhombic $CeNiSi_2$-type layered structure.[4] For polycrystalline samples of $CeNiGe_2$, the Sommerfeld coefficient of $\gamma = 220$ mJ/K^{2}mol was found.[5] The magnetic susceptibility and specific heat measurements indicated two antiferromagnetic phase transitions at $T_N^{I} = 4$ K and $T_N^{II} = 3$ K.[6] However, the nature of two-step antiferromagnetic ordering below 4 K has not yet been elucidated. Furthermore, one can expect anisotropic properties in response to the direction of current and magnetic field along and perpendicular to the plane.

We have grown single crystals of $CeNiGe_2$ by using a Sn-flux method, from which we obtained many plate-like single crystals of the size approximately $1 \times 3 \times 3$ mm^3 oriented along the b axis. Powder x-ray diffraction pattern reveals that $CeNiGe_2$ crystallizes in the orthorhombic $CeNiSi_2$-type (space group Cmca) structure. The lattice parameters are $a = 4.25(3)$ Å, $b = 16.78(9)$ Å, and $c = 4.21(0)$ Å, close to those reported previously.[4] The in-plane resistivity was measured on a plate-like sample by a standard four-probe AC method, while the b-axis resistivity was estimated from the measurements using a modified four-probe configuration, as used in quasi-2D crystals.[7, 8] Magnetization for $H//b$ and $H\perp b$ and specific heat in zero field were taken using a Quantum Design SQUID magnetometer and a Quantum Design PPMS system, respectively.

Figure 1 displays the anisotropic behavior of the magnetization $M(H)$ of $CeNiGe_2$, which was measured at 2 and 3.5 K below and between the two transition temperatures in magnetic fields to 5 T applied along the b axis ($H//b$) and in the plane ($H\perp b$). There is a very large anisotropy with an easy magnetization direction along the b axis. $M(H//b)$ shows a metamagnetic-like behavior around 0.7 T and then saturates rapidly to a value of 1.2 μ_B/Ce. The high-field measurement indicated that $M(H//b)$ at 500mK reaches 2 μ_B/Ce at 50 T. On the other hand, $M(H\perp b)$ increases linearly with increasing magnetic field. At $H = 5$ T, the magnetization ratio of $M(H//b)/M(H\perp b)$ is estimated to be about 15. This large anisotropy in response to the magnetic field is further confirmed by the measurements of magnetic susceptibility.

The magnetic susceptibility $\chi(T)$ and its inverse $\chi^{-1}(T)$ in a field of 0.1 T parallel ($H//b$) and perpendicular to the b axis ($H\perp b$) as a function of temperature from 2 to 350 K are shown in Fig. 2. As expected from the $M(H)$ observations, the b-axis magnetic susceptibility $\chi_{//b}$ is much larger than the in-plane magnetic susceptibility $\chi_{\perp b}$ over all temperature range. Above 100 K, the data obey the Curie-Weiss law, $\chi = C/(T-\theta_p)$ with the paramagnetic Curie temperatures of $\theta_{//b} = 31.9$ K and $\theta_{\perp b} = -168.2$ K. This might indicate a development of ferromagnetic and antiferromagnetic exchange interactions for $H//b$ and $H\perp b$, respectively. From the value of $\theta_p = (\theta_{//b} + 2\theta_{\perp b})/3$, we can calculate the

Kondo temperature $T_K \sim |\theta_p/2|$,[9] obtaining $T_K = 50$ K. The high-temperature slopes of χ^{-1} yield the effective magnetic moments of $\mu_{//b} = 2.11\ \mu_B$ and $\mu_{\perp b} = 2.22\ \mu_B$, which is slightly smaller than the theoretical value (2.54 μ_B) expected for the free Ce^{3+} ion. This implies that the magnetic moments of Ce ions are well localized in this material. The deviation from the Curie-Weiss behavior below 100 K could be attributed to the crystal field effect. Inelastic neutron scattering studies are further required in order to understand the crystal field states. The low-temperature data of $\chi_{//b}$ and $\chi_{\perp b}$ exhibit two anomalies at 4 and 3 K indicating antiferromagnetic orderings. Figure 3 shows up the low-temperature data of the ratio of magnetic susceptibility $\chi_{//b}/\chi_{\perp b}$, specific heat divided by the temperature C/T, and the temperature derivative of in-plane resistivity $d\rho_{\perp b}/dT$. There are obvious two magnetic transitions at 4 and 3 K, being consistent with previous works for polycrystalline samples.[6]

In Fig. 4, we have plotted the in-plane and b-axis resistivity curves as a function of logarithmic scale of the temperature. The in-plane resistivity $\rho_{\perp b}(T)$ is characterized by a broad peak around 100 K, followed by a minimum at ~ 20 K, and a steep decrease below 4 K. The peak structure is typical of that expected when there is an influence of the crystal field on Kondo effect.[10] The steep decrease below 4 K could be attributed to the combined effect of the reduction of spin-disorder scattering and the development of coherence, as found in other Kondo antiferromagnetic compounds.[2, 3, 11] The difference between $\rho_{//b}(T)$ and $\rho_{\perp b}(T)$ is likely to result from different types of hybridization between localized $4f$ electrons and conduction electrons between the b-axis and in-plane and/or lattice disorder in the plane. On cooling from 200 to 2 K, the resistivity ratio $\rho_{//b}/\rho_{\perp b}$

decreases from 65 to 5. This anisotropic transport is suggestive of dominant motion of conduction electrons in the plane.

In summary, we have measured the magnetization, magnetic susceptibility, specific heat, and electrical resistivity on a single crystal of $CeNiGe_2$. This material is a Kondo lattice compound undergoing two antiferromagnetic transitions at 4 and 3 K. The easy magnetization axis is parallel to the longest crystallographic b direction. The magnetization ratio of $M(H//b)/M(H\perp b)$ is estimated to be about 15 in a field of 5 T. The conductivity is dominated in the plane. The resistivity ratio of $\rho_{//b}/\rho_{\perp b}$ increases from 5 to 65 on heating from 2 to 200 K. Therefore, $CeNiGe_2$ is classified into a Kondo antiferromagnet with a strong anisotropy arising from the quasi-2D crystal structure.

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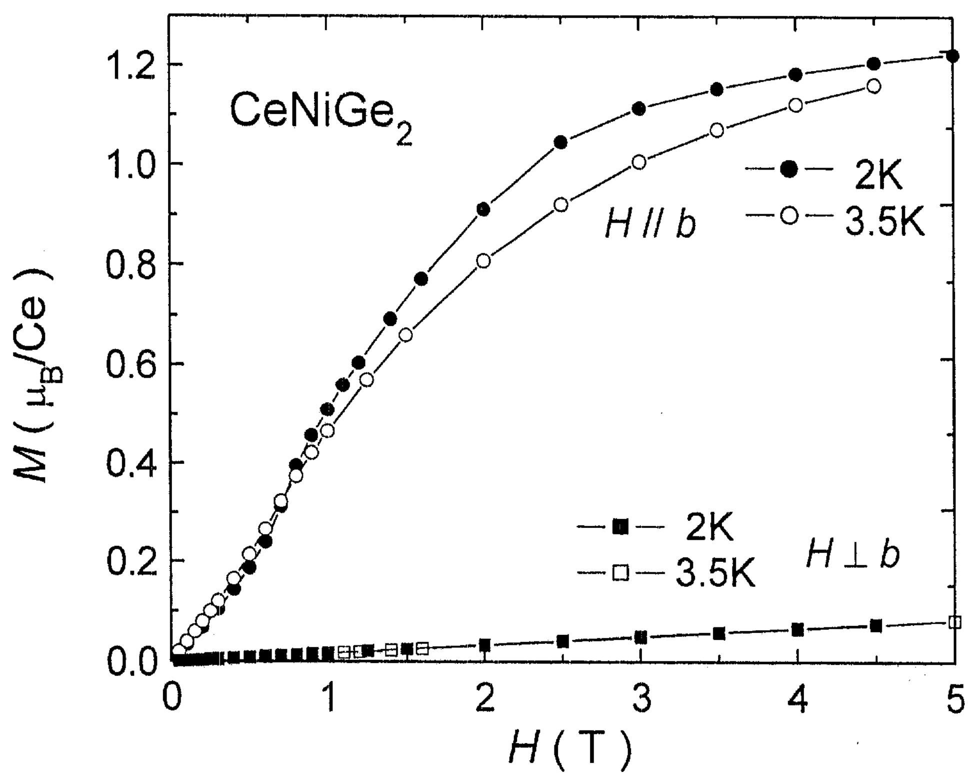
Figure captions

Fig. 1. Isothermal magnetization $M(H)$ versus applied field to 5T for $CeNiGe_2$ single crystal at temperatures 2 and 3.5K for $H//b$ and $H \perp b$.

Fig. 2. Magnetic susceptibility χ and χ^{-1} versus temperature between 2 and 350K for $CeNiGe_2$ in an applied magnetic field of 0.1T for $H//b$ and $H \perp b$.

Fig. 3. Low-temperature data of (a) magnetic susceptibility ratio $\chi_{//b}/\chi_{\perp b}$, (b) specific heat divided by the temperature C/T, and (3) temperature derivative of the in-plane resistivity $d\rho_{\perp b}/dT$ for $CeNiGe_2$.

Fig. 4. In-plane resistivity $\rho_{\perp b}(T)$ and b-axis resistivity $\rho_{//b}(T)$ for $CeNiGe_2$ as a function of temperature.



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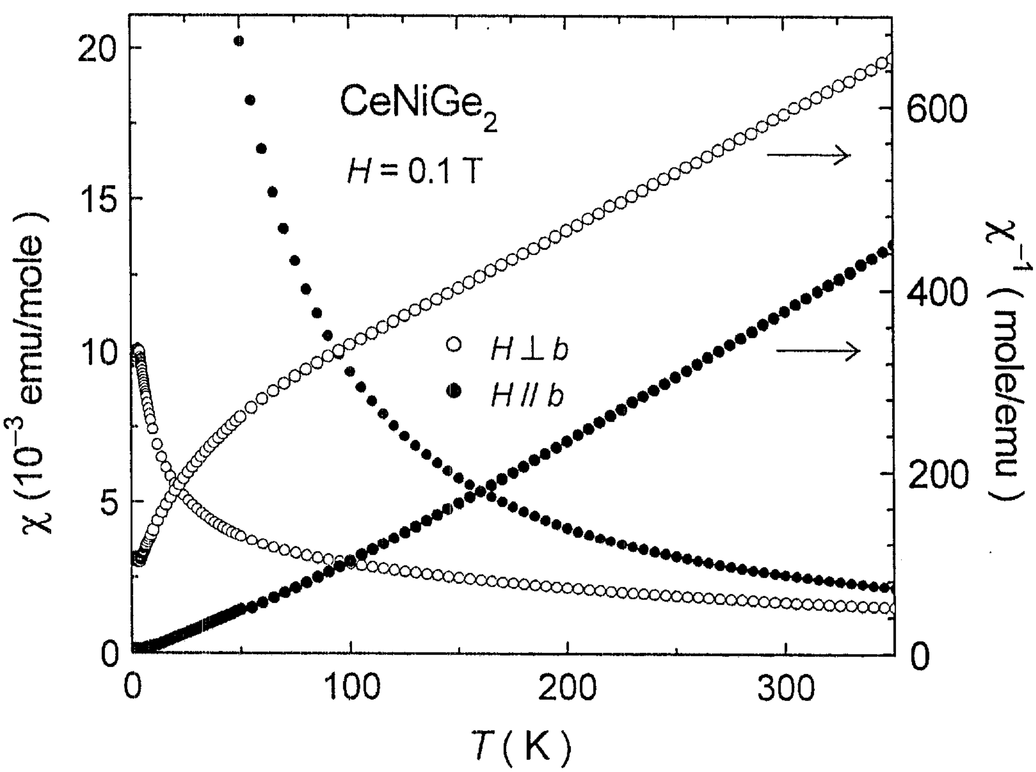

Fig. 2. M. H. Jung et al.

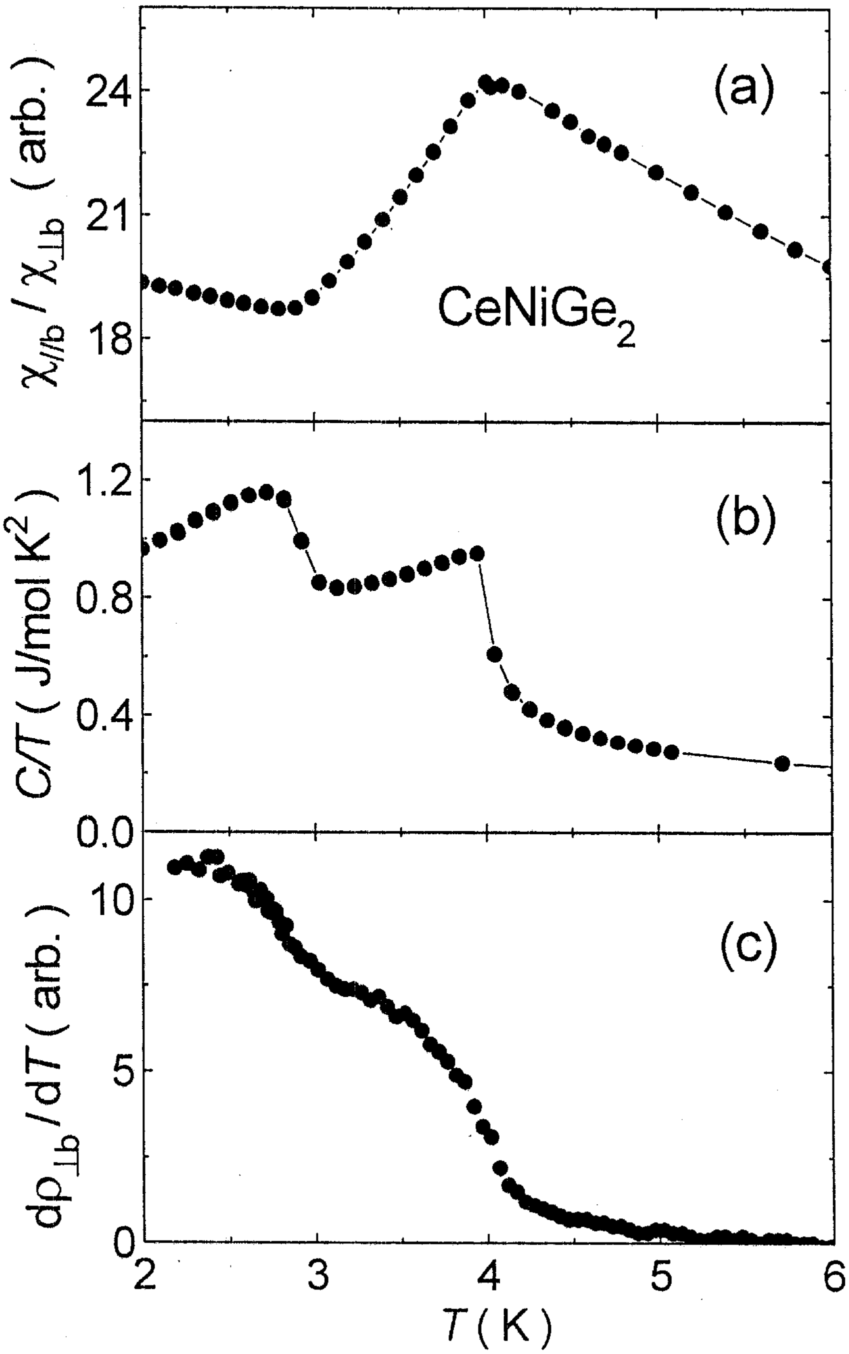

Fig. 3. M. H. Jung et al.

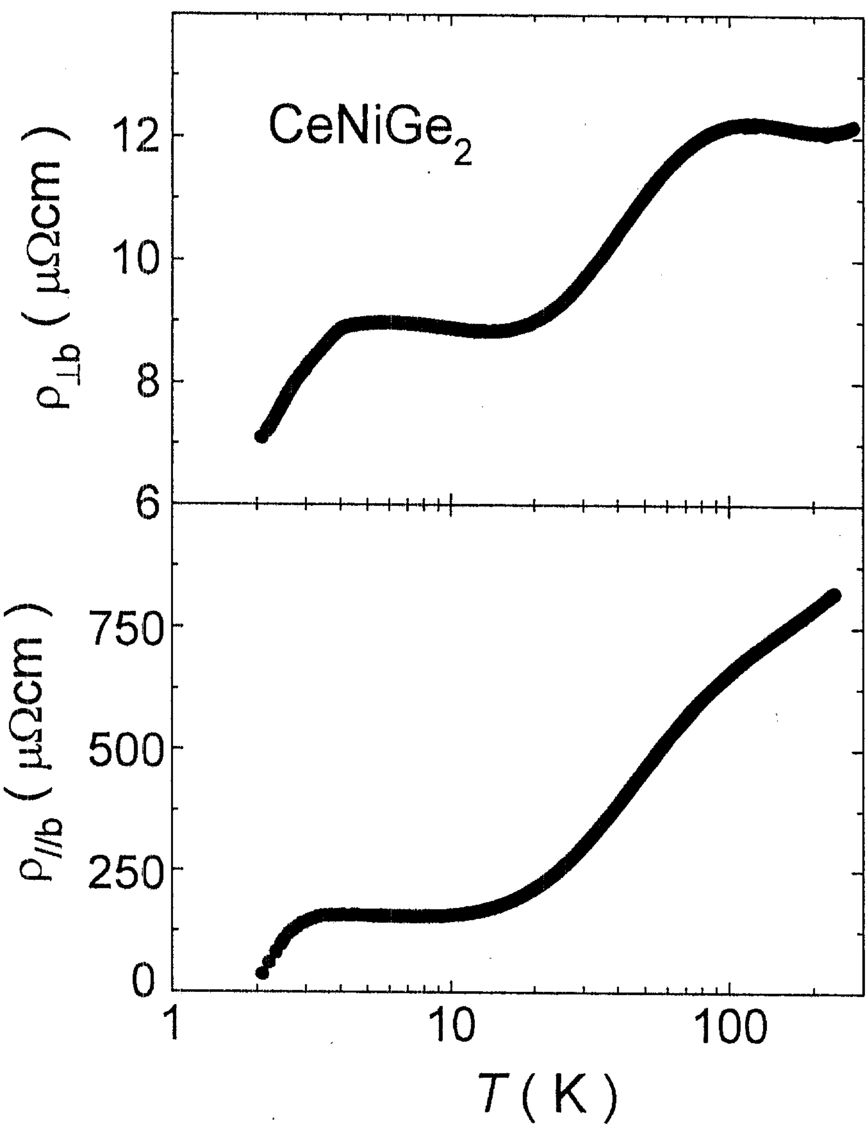

Fig. 4. M. H. Jung et al.